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RESEARCH ESTABLISHMENT
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THE DETECTION AND ESTIMATION OF INDIUM-114m IN EFFLUENT

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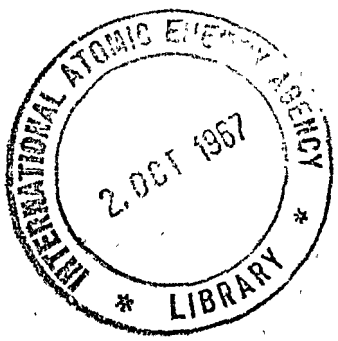

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ABSTRACT

A simple method for the quantitative determination of indium-114m in effluent is described. Five litres of effluent is scavenged with ferric hydroxide and the indium-114m determined by gamma spectrometry. The method is rapid, economical of operator time and is applicable down to concentrations of $2 \times 10^{-4} \mu Ci/\ell$ and lower.

CONTENTS

1. INTRODUCTION

A method for monitoring In-114m in the effluent from the A.A.E.C. Research Establishment is required as this nuclide has one of the lowest allowable concentrations. Since it is expected that In-114m will not occur frequently in the effluent, a quick method, or one involving little attention, is preferred. This report describes a method involving only a ferric hydroxide scavenge of five litres of effluent, separation, then subsequent gamma spectrometry of the precipitate.

2. EXPERIMENTAL

2.1 Preparation of Standard In-114m

A solution of In-114m in N/100 HCl was prepared and standardised by $4\pi\beta$-counting of the In-114 daughter. In-114 has a half life of approximately 70 seconds and is thus in secular equilibrium with In-114m. 96.5 per cent. of In-114m decays to In-114 by isomeric transition and the remainder by electron capture to Cd-114. 98 per cent. of In-114 decays by β^- emission to Sn-114 and the remainder by electron capture and β^+ emission (negligible) to Cd-114. Corrections for the branching ratios were made to the $4\pi\beta$ count to obtain the In-114m disintegration rate.

2.2 Sensitivity of Detection of In-114m by Gamma Spectrometry

An aliquot of standard In-114m, containing approximately $4.28 \times 10^{-4}\mu$Ci, an amount equivalent to one-fifth of the maximum permissible discharge level in one litre of effluent, was taken and counted by γ spectrometry on a 3 in. x 3 in. NaI(Tl) crystal for 40 minutes and the background subtracted. The spectrometer was set up so that the 0.192 MeV γ emitted, which is the most abundant in In-114m, would occur at about the centre of the spectrum. This resulted in a spectrum in which the 0.192 MeV peak could just be seen but with no great certainty.

The experiment was repeated using five times the above quantity of In-114m. In the resultant spectrum, the 0.192 MeV peak was easily detectable above the scattered points produced by accumulation and subtraction of background. This spectrum is shown in Figure 1.

2.3 Concentration of Indium Activity in Effluent

Since five litres of effluent is an inconveniently large sample for γ spectrometry, a method of concetrating any indium activity present was required. Ferric hydroxide, which Sunderman (1956) found would carry down over 99.7 per

cent. of active indium from solution under carrier-free conditions and in the presence of inactive indium, was chosen as the scavenging agent.

The most suitable method for separating the ferric hydroxide precipitate and other solid material in the sample from the liquid was as follows:

Five litres of effluent containing the ferric hydroxide precipitate was allowed to stand overnight in a container, preferably one with vertical sides to allow the precipitate to settle. As much of the supernatant liquid as possible was withdrawn without disturbing the precipitate, using as filter stick a cut down two inch diameter Buchner funnel fitted with a Whatman No. 1 filter paper. A water vacuum pump used with compressed air instead of water provided sufficient vacuum for this purpose. The remaining sample was transferred to a one litre measuring cylinder, allowed to settle for one to two hours and as much of the supernatant liquid as possible was withdrawn as before.

Using this method the volume can be reduced from five litres to 100-150 ml in a few hours with very little operator attention.

2.4 Preparation of Blank Effluent Sample

A source was prepared from five litres of effluent by scavenging with ferric hydroxide and was counted for 1000 minutes with a 3 in. x 3 in. NaI(Tl) crystal using a 256 channel analyser. The spectrum obtained is shown in Figure 2. No γ peaks can be seen in the 0.2 MeV region.

2.5 Sensitivity of Detection of In-114m in Effluent

Five litres of the same effluent used in the preparation of the blank sample was spiked with 3.8×10^{-4} µCi of In-114m. This is equivalent to 0.082 of the maximum permissible monthly discharge level. The solution was scavenged with ferric hydroxide and a source was prepared and counted with the 3 in. x 3 in. crystal γ spectrometer for 1000 minutes.

The spectrum obtained is shown in Figure 3. The 0.192 MeV peak due to In-114m is clearly visible, showing that less than one tenth of the maximum permissible monthly discharge level can be detected by this method.

2.6 Recovery of In-114m

Five litres of the same effluent as used before was spiked with 1.03×10^{-2} µCi of In-114m and scavenged with ferric hydroxide. A source was prepared from the precipitate.

A standard source was prepared from 1.03×10^{-2}µCi of In-114m.

Both sources were counted twice on a 5 in. x 4 in. NaI(TI) γ spectrometer for forty minutes and the background subtracted. The 0.192 MeV peak obtained in each count was integrated, corrected for Compton scatter contribution from γ peaks of higher energy and the recovery of In-114m was calculated. The results obtained are given in Table 1.

The spectrum obtained from one of the spiked effluent counts is shown in Figure 4.

TABLE 1

Recoveries of In-114m

Spiked sample first count/Standard first count	97.4%
Spiked sample first count/Standard second count	90.3%
Spiked sample second count/Standard first count	111.9%
Spiked sample second count/Standard second count	103.6%
mean	100.8%

3. PROCEDURE

3.1 Preparation of Source

Place five litres of effluent in a two gallon container. Acidify with dilute (1:1) HCl add 1g of $Fe(NO_3)_3.6H_2O$, dissolve and precipitate by adding excess NH_4OH. Allow to stand overnight and reduce volume to 100-150 ml as described in Section 2.3. Remove the remainder of the supernatant liquid by centrifuging. Transfer precipitate to a 100 ml beaker with water, and heat under an infra-red lamp until most of the clear liquid above the solid is removed but do not allow the solid to dry out. Add the filter paper used in the filtration of the bulk to the beaker.

3.2 Counting

Count the sample source on a 3 in. x 3 in. NaI(Tl) γ spectrometer for 1000 mins. If no peak is detected at 0.192 MeV report In-114m as less than 1.8×10^{-4}µCi/ℓ. If a peak is detected, count a standard source under the same con-

ditions as the sample source.

3.3 Preparation of Standard Source

Place a known aliquot of a solution of In-114m standardised by 4πβ counting in a similar 100 ml beaker to that used in preparing the sample source. Dilute with water to approximately the same volume as the precipitate in the sample source.

3.4 Calculation of Result

Determine the width of the In-114m peak from the standard spectrum. In this study it was ± 16 channels from the peak and this figure is used as an example.

Determine the Compton scatter from other emitters in the sample and the background contributions by taking the mean of the counts in channels maximum + 16 and maximum -16 and multiply by 33 (number of channels in the peak).

Integrate the channels in the sample peak and subtract the correction figure obtained above.

Treat the standard spectrum in the same manner making a correction in a similar manner to that used for the correction of the sample count.

Then:

$$\mu\text{Ci In-114m}/\ell \text{ of sample} = \frac{\text{corrected count (sample)} \times \mu\text{Ci In-114m (standard)}}{\text{corrected count (standard)} \times 5}$$

4. INTERFERENCES

Interference in this method would be caused by any nuclide having a γ peak from 0.18 - 0.20 MeV which is carried by ferric hydroxide. The nuclides listed by Crouthamel (1960) having γ peaks in this region and half-lives over one hour, formed by fission or n, γ reactions, are given in Table 2. All can be identified by other peaks in their γ spectra. Actual spectra plots for some of these are given by Crouthamel (1960) or Heath (1964) and the spectra of the rest can be obtained from information given by Strominger et al. (1958).

TABLE 2

INTERFERING NUCLIDES WITH HALF-LIVES OVER ONE HOUR

Nuclide	Half-Life	Spectrum Reference
Mo-99/Tc-99m	2.75d.	Crouthamel, Heath
Te-131m	1.2d.	Strominger et al.
Dy-157	8.2h.	Strominger et al.
Ho-166m	30y.	Heath
Sn-117m	14d.	Crouthamel
Hf-181	44.6d.	Crouthamel, Heath
Pt-197	17.4h.	Crouthamel, Heath
Fe-59	45.1d.	Crouthamel, Heath
Hg-197	2.7d.	Heath
Yb-169	31.8d.	Crouthamel, Heath
Ge-75	1.4h.	Crouthamel, Heath
Eu-156	15.4d.	Heath

If the In-114m result obtained is sufficiently high to prevent the discharge of the effluent, the presence of In-114m should be confirmed by locating its 0.552 MeV and 0.722 MeV peaks and comparing the three peak ratios. A spectrum of pure In-114m showing these peaks obtained with a 3 in. x 3 in. NaI(Tl) crystal is given in Figure 5.

5. CONCLUSION

The method described is simple and requires little operator time. It will easily detect less than one-tenth of the maximum permissible monthly discharge level of In-114m.

6. ACKNOWLEDGEMENT

The assistance of Mr. A. R. Palmer is gratefully acknowledged.

7. REFERENCES

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Crouthamel, C. E. (Editor) (1960). - Applied Gamma Spectrometry. Pergamon Press.

Heath, R. L. (1964). - Scintillation Spectrometry IDO 16880. Pt. 2.

Strominger, D., Hollander, J. M., Seaborg, G. T. (1958). - Reviews of Modern Physics 30: (2) Part II.

APPENDIX I

MAXIMUM PERMISSIBLE MONTHLY DISCHARGE OF IN-114m

$= 29\ \text{mCi}/(3 \times 10^{6})\ \text{gallons}$

$= 4750\ \text{d.p.m.}/\ell$

$= 2.14 \times 10^{-3}\ \mu\text{Ci}/\ell$

$= 2.14 \times 10^{-6}\ \mu\text{Ci}/\text{m}\ell$

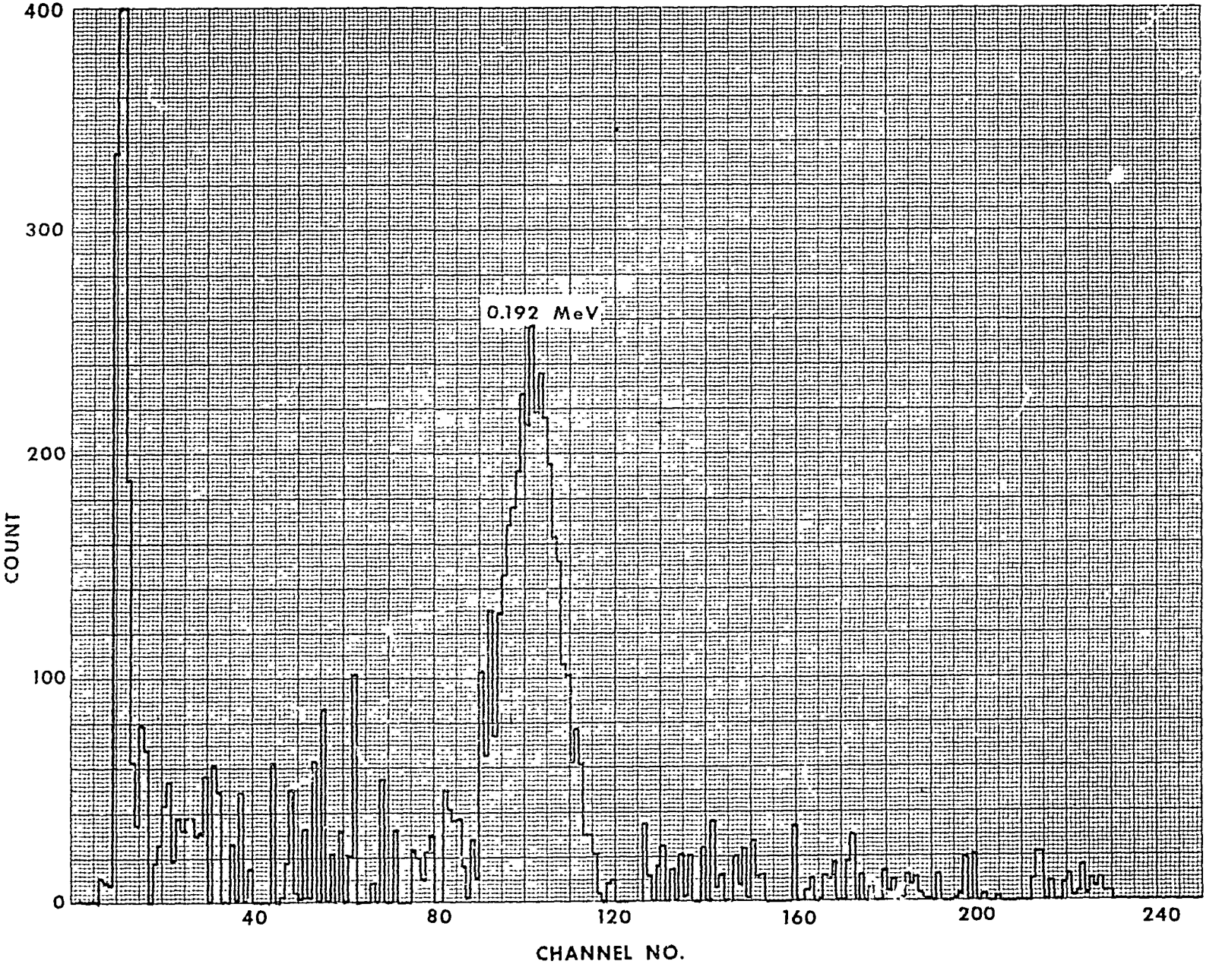

FIGURE 1. γ SPECTRUM OF 2×10^{-3} μCi INDIUM-114m

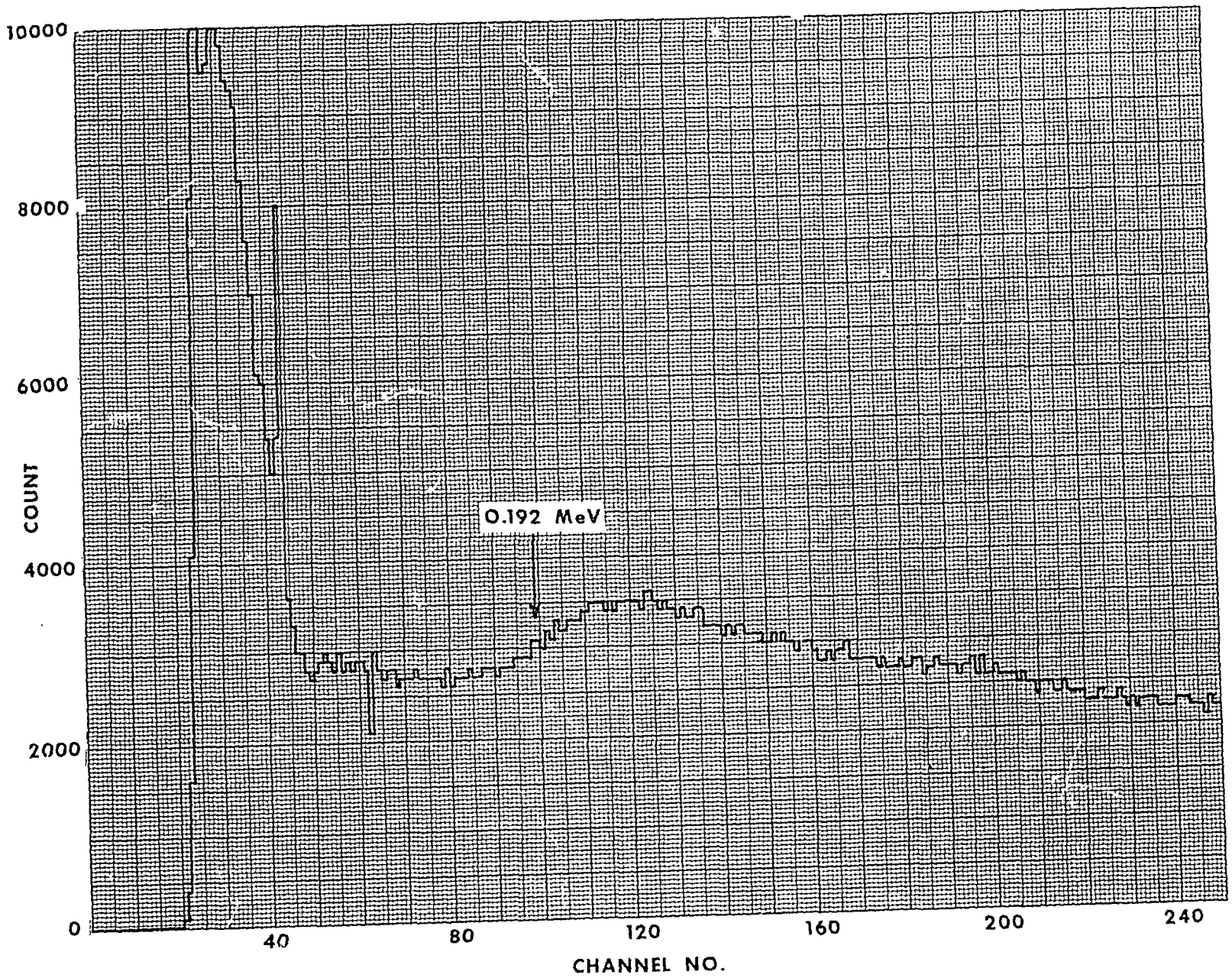

FIGURE 2. γ SPECTRUM OF UNSPIKED EFFLUENT

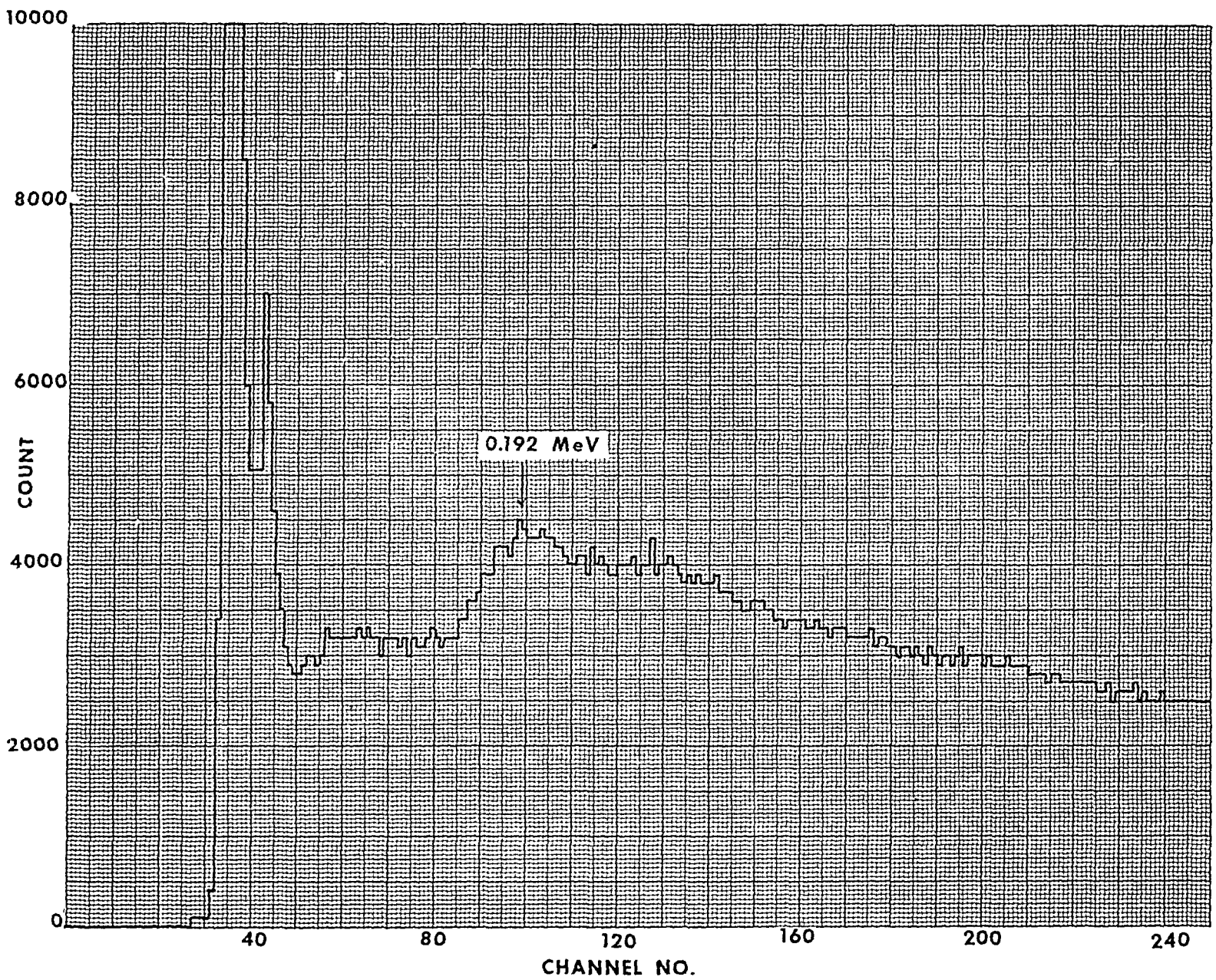

FIGURE 3. γ SPECTRUM OF EFFLUENT SPIKED WITH 8.8×10^{-4} μCi INDIUM-114m

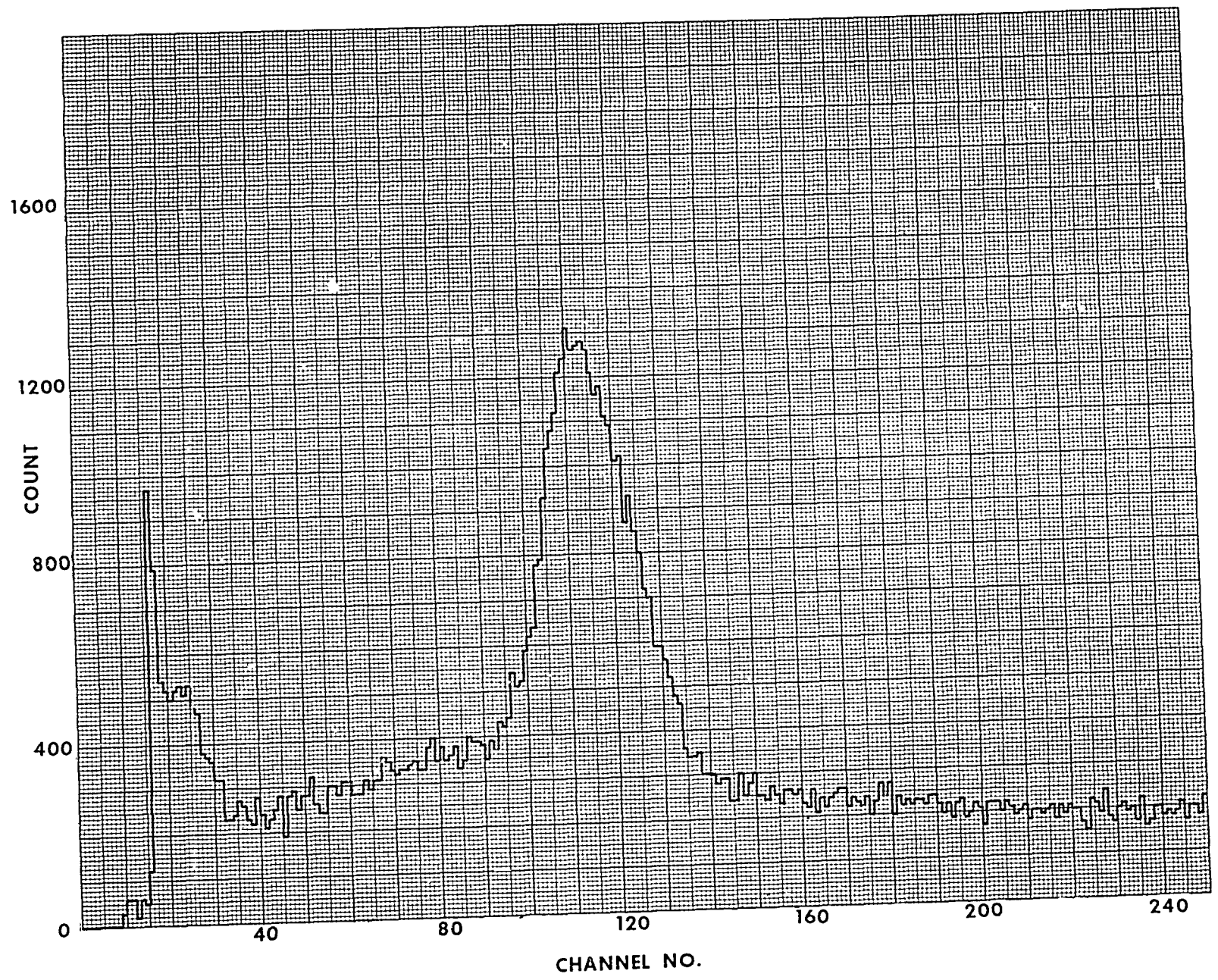

FIGURE 4. γ SPECTRUM OF EFFLUENT SPIKED WITH 1.03×10^{-2} μCi INDIUM-114m

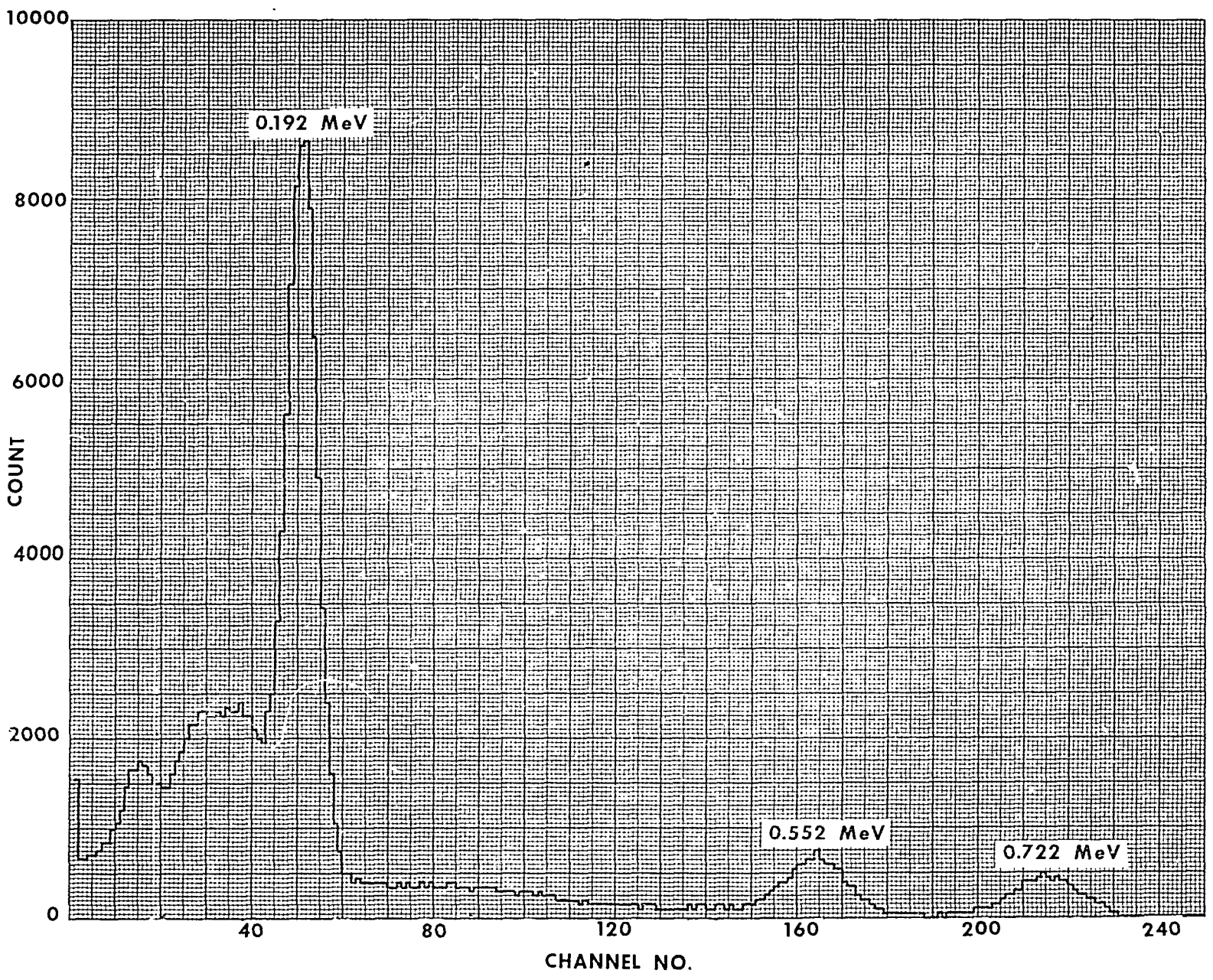

FIGURE 5. γ SPECTRUM OF INDIUM-114m OBTAINED WITH 3in.×3in. NaI(Tl) CRYSTAL